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QUARTERLY REPORT
for
DARPA/ONR
HIGH TEMPERATURE SUPERCONDUCTIVITY

PERIOD ENDING: March 31, 1990

I. PROGRAM INFORMATION

Contract Number:	N00014-88-C-0760
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II. PROGRAM SUMMARY

The overall goals of this program are to develop the technology of MBE growth of HTSC material, to optimize the performance of HTSC films with high transition temperatures and critical current densities, and to explore the development of electronic devices based on such material.

III. PROGRAM STATUS

The recently developed MBE system is functioning well and superconducting films with T_c's above 77K are now routinely being grown, in-situ, by atomic layer epitaxy on both MgO and $SrTiO_3$. Work is continuing on understanding such issues as required stoichiometric control and methods of improving kinetic control precision, oxidation capacities of reactive oxygen sources, and the effect of crystallographic quality on superconducting properties.

IV ACCOMPLISHMENTS

Work has continued on studying the growth of layered superconductors using atomically-layered epitaxy. An incremental improvement in the measured transition temperature was obtained, from 84 to 86K. This was from a nominal 2223-phase film. In addition, these films remain single crystal heteroepitaxial with isolated defects now observed as low as 10^3 cm^{-2}. The improved defect density seems to be correlated with improved stoichiometric control from careful and sustained calibration analysis.

With the ability to grow single-crystal films that appear to be atomically smooth, it is interesting to consider the growth of layered artificial structures, i.e., films whose crystallographic nature is changed on a unit cell-by-unit-cell basis. Ultimately, such structures as layered S-I-S or S-Se-S or S-S'-S films may need to be grown with

atomic layer precision and sharp crystallographic interfaces for advanced junction devices. To demonstrate the capability of well-calibrated, atomically-layered epitaxy, we studied the growth of superlattice structures where the compound being grown changes each unit cell.

The family of compounds $Bi_2Sr_2Ca_{n-1}Ca_nO_x$ contains members that differ in the number of CuO_2 layers, and hence the thickness of the peroskite-like subcell. We have been able to grow single-phase films of all of the compounds containing from one to five CuO_2 layers per unit cell. X-ray diffraction patterns from these are shown in Fig. 1. These compounds provide a good set of materials for studying the growth of superlattice structures. It can be shown that the X-ray diffraction patterns from the superlattices, consisting of alternating layer of say the 2212 and 2223 compounds, contain twice the number of peaks in a normal c-axis scan as does a similar scan of a film consisting of a single phase. Due to the order inherent in the superlattice, however, one half of the peaks are substantially suppressed in amplitude. In addition, for superlattices of unit cells with similar structure factors, this peak suppression is very strong; the more similar the structure factor, S(k), is for a given k(2θ), the more complete the suppression of half the peaks is. Furthermore, it can be shown that for small 2θ angles, i.e., small values of k, the odd peaks are suppressed, while for somewhat larger values of 2θ, the even reflection peaks are suppressed. For still larger values of 2θ again, the odd reflection peaks are again suppressed. This pattern continues with alternately the odd indexed peaks or the even indexed peaks suppressed due to the regular structure of the superlattice.

A superlattice consisting of alternating 2212 and 2223 unit cells was grown by atomically-layered epitaxy on $SrTiO_3$. This was accomplished by programming the computer which controls the beam shuttering times to grow alternating layers of 2212 or 2223 unit cells. The XRD pattern of the resulting film is shown in Fig. 2. All of the peaks can be indexed to a lattice constant of about 34 Å,

the sum of the length of a compositional 2212 and 2223 unit cells. Note that both odd and even indices need to be employed to account for all of the peaks, and that for intermediate angles, ~30°, the even peaks are suppressed; while for lower and higher angles, the odd peaks are suppressed. That is, indeed, what the simple diffraction model described above predicts.

It is useful to note that a closely related analysis also predicts diffraction patterns that appear to consist of both even and odd indexed peaks. This is the fluctuation diffraction analysis of Hendricks and Teller (1942). They describe diffraction patterns of random mixtures of two unit cells. In particular, for equal quantities of the two unit cells, a diffraction pattern with peaks at positions close to the main ones (i.e., those not suppressed) shown in Fig. 2 is predicted. However, no peaks occur at the angles of the suppressed peaks. Thus the observation of the suppressed peaks is a good indication that the structure consists of a regular, alternating sequence, i.e., a superlattice, of the two unit cells and not a random mixture.

In particular, for random mixtures, Hendricks and Teller's theory says that no peak should be found at an angle corresponding to a lattice spacing equaling the sum of the two unit cell values, while superlatttice diffraction theory predicts a suppressed peak (the 001 peak) to occur there. The 001 peak is an unambiguous indicator that the film consists of alternating superlattice layers.

We have studied low-angle diffraction from several of our superlattice films. The 001 peak consistently appears at an angle corresponding to the sum of the two unit cell c-axis lattice constant values. Figure 3 shows such a scan along with two theoretical predictions for a case when the two unit cells were the 2201 and 2223 phase. Since the unit cells differ substantially in the Fourier transform of their structure factors at small angles, the suppression of the 001 peak is small and the peak shows up strongly. The second curve shows a predicted diffraction pattern computed numerically

for this film. Note the excellent agreement. On the other hand, the fluctuation diffraction theory of Hendricks and Teller predicts the curve shown below. This result clearly demonstrates the ability of atomically-layered epitaxy to grow modulated heterostructures with monolayer precision.

V PROBLEM AREAS

No specific problem areas exist at the present time. More film growth is required to improve film properties and attempt the fabrication of metastable structures.

VI CORRECTIVE ACTION

None required at present

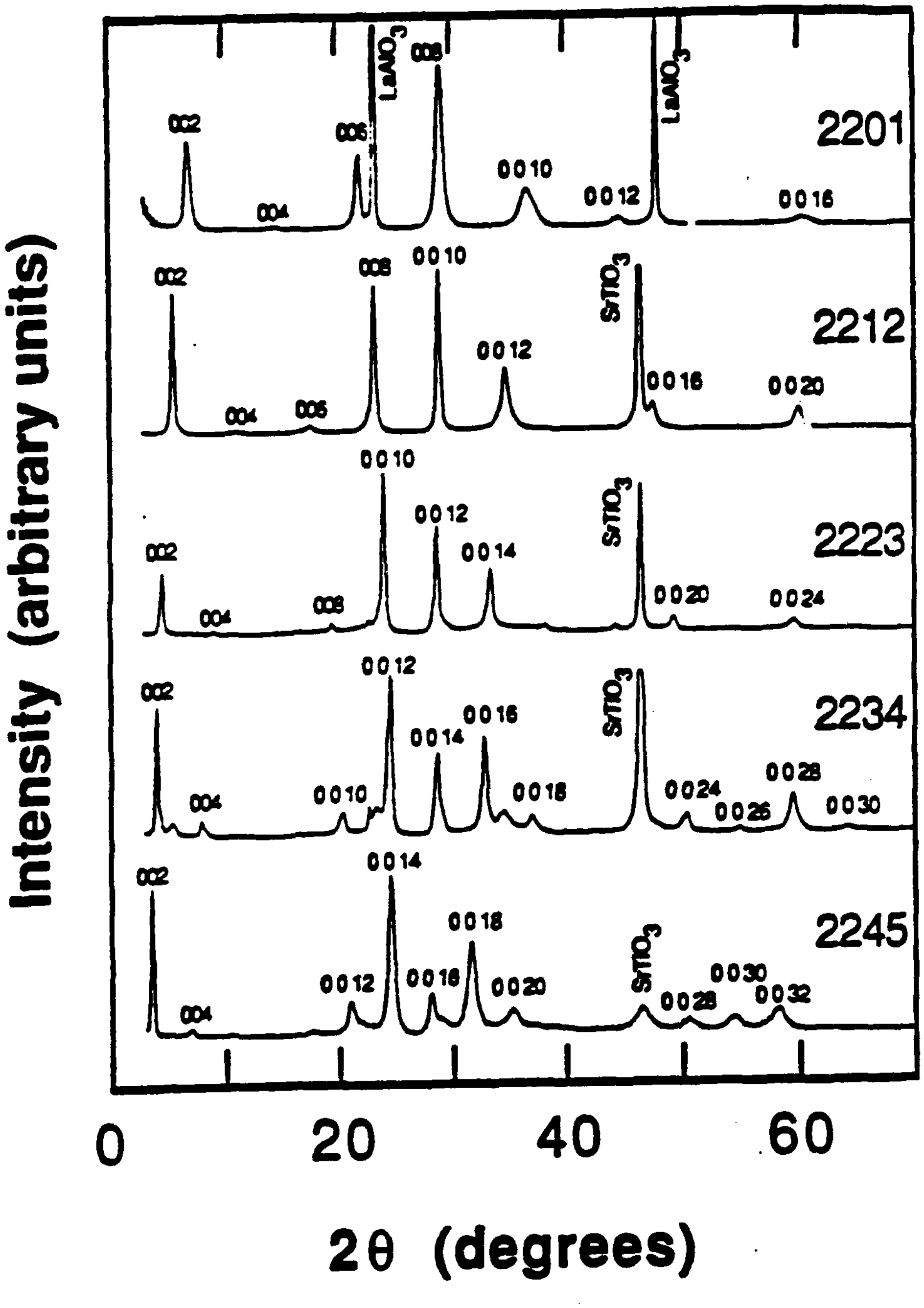

Fig. 1 X-ray diffraction patterns obtained from five different growths in which compounds containing from one to five CuO_2 layers per unit cell were grown.

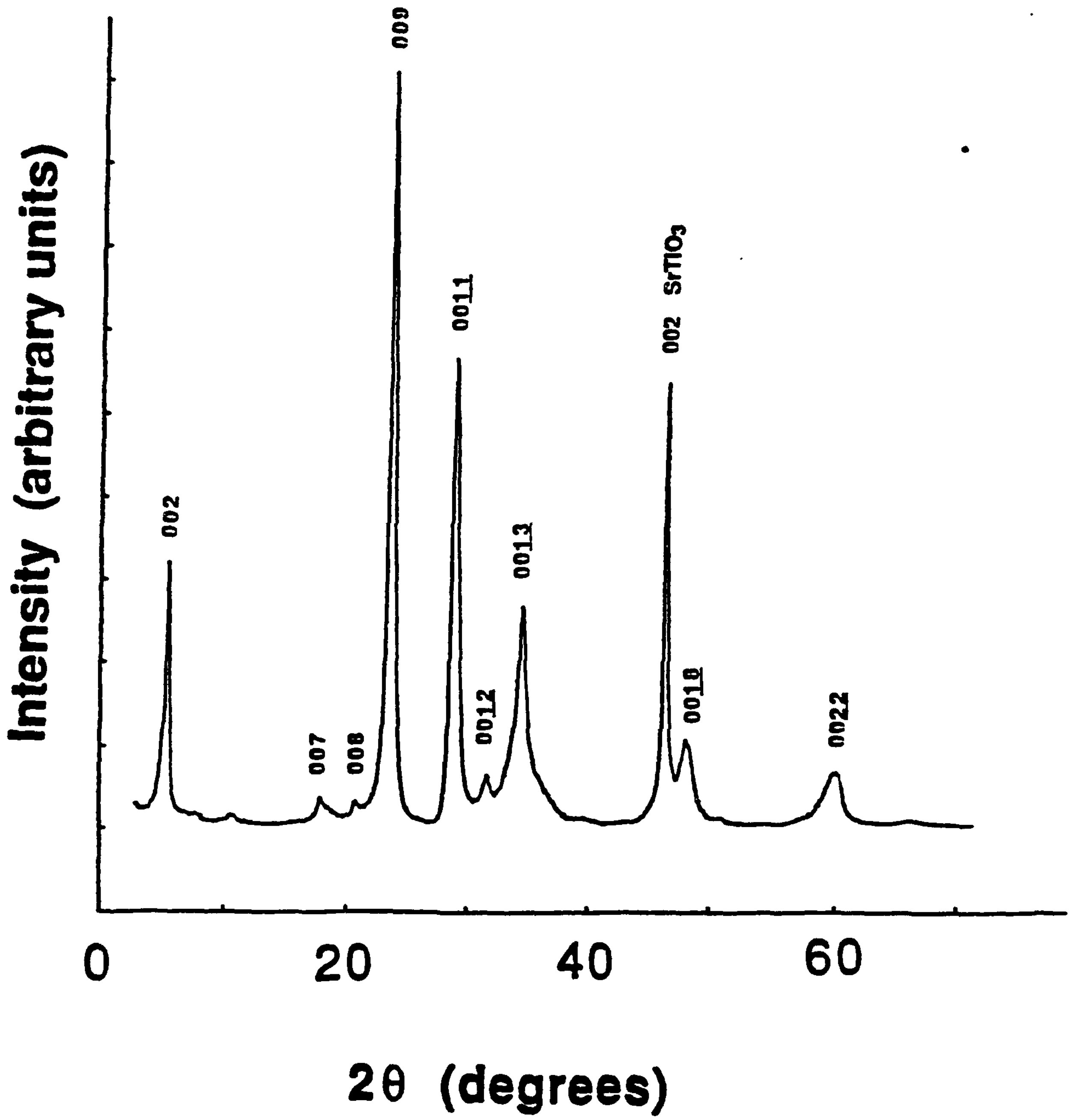

Fig. 2 X-ray diffraction pattern of superlattice consisting of alternating unit cells of the 2212 and 2223 compounds.

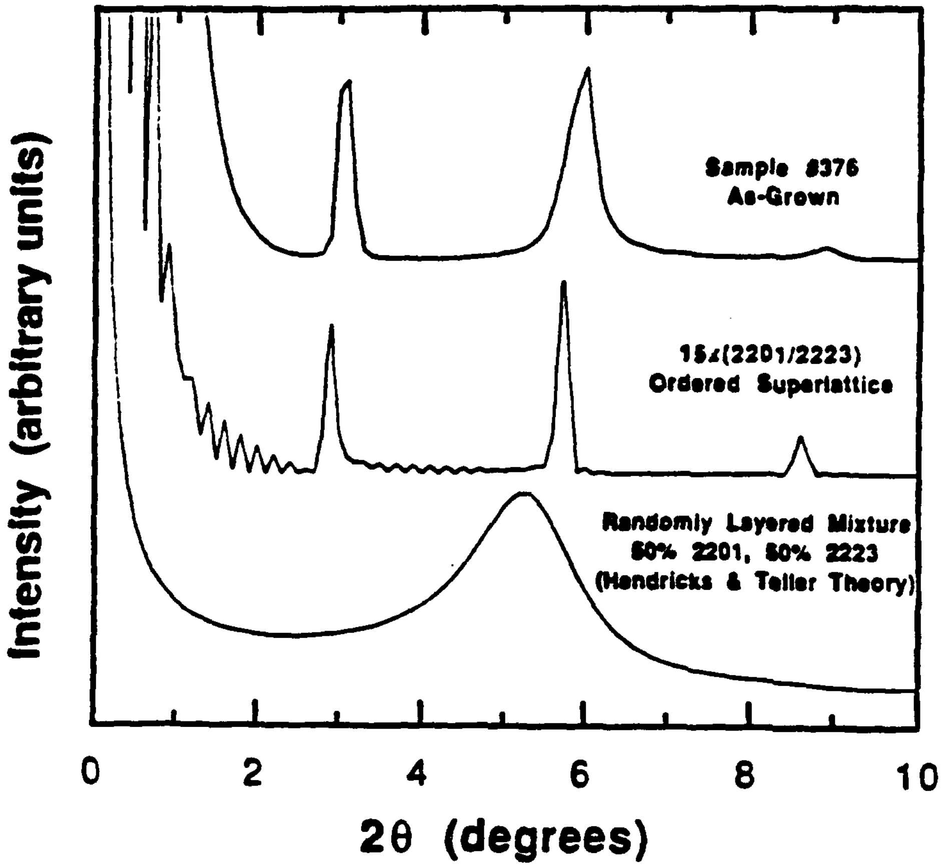

Fig. 3 Low angle x-ray diffraction pattern obtained from a superlattice film consisting of alternating unit cells of the 2201 and 2223 compounds. Also shown are theoretical calculations for a 15 period 2201/2223 superlattice and the predictions of Hendricks and Teller's theory for a film consisting of a random mixture of 2201 and 2223 unit cells.